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CLAIMS

[Claim(s)]

1. Approach which is approach of manufacturing 2-n-butyl-2-ethyl-1,3-propanediol from 2-ethyl HEKIZANARU and formaldehyde under existence of basic alkali-metal hydroxide catalyst or alkaline earth metal hydroxide catalyst, and is formalin solution with which source of formaldehyde used contains solid-state paraformaldehyde and ** methanol at less than 2 % of the weight, and is characterized by being used in order that further cationic or neutral phase transition catalyst may increase reaction.
2. Approach characterized by using neutral or cationic phase transition catalyst in approach of said claim 1 when source of formaldehyde is content **** formalin solution at less than 1 % of the weight about methanol.
3. Approach characterized by using cationic phase transition catalyst in approach of said claim 1 when it is source of formaldehyde, or solid-state paraformaldehyde.
4. Approach characterized by neutral phase transition catalyst being polyethylene lender recall in approach of said claim 2.
5. Approach characterized by cationic phase transition catalyst used being TORIKA prill, methylammonium chloride or sulfuric-acid acidity quarternary ammonium salt in approach of said claims 2 or 3.
6. Formalin solution used as source of formaldehyde in said claim 1, or approach characterized by containing methanol at less than 1 % of the weight.
7. Approach characterized by consisting of the following synthetic phases performed [whether PUROSESE made it the following Sequence A or Sequence B and] in approach of said claim 1 for manufacture of 2-n-butyl-2-ethyl-1,3-propanediol.
 - A) 1) It acts as the ** people of formaldehyde, 2-ethyl hexanal, and the cationic or neutral phase transition catalyst into a reactor, and subsequently to 30-100 degrees C mixture is heated, 2) The water solution of an alkali hydroxide or an alkaline earth hydroxide is added by said mixture at ****, and stirring is continued over 0 - 3 hours at said temperature, 3) That said mixture is neutralized by the mineral acid or the organic acid, a phase is separated, and an organic phase is washed with water, that vacuum distillation of the 4 organic phase is carried out by 130-132 degrees C / 9mmHg, or B
 - 1) Formaldehyde, an alkali-metal hydroxide or an alkaline earth metal hydroxide, and a cationic or neutral phase transition catalyst are inserted into a reactor, and the mixture is heated by 30 to 100 degree C, 2) It applies for 0 to 5 hours, and it is dropped into the mixture, 2-ethyl hexanal is added, and churning is continued at the temperature for 0 to 3 hours, 3) Phase separation is neutralized and carried out by the mixture, the mineral acid, or the organic acid, and an organic phase is washed with water, 4) Carrying-out [vacuum distillation of the organic phase]-by 130-132-degree-C / 9mmHg 8. In one process of said claims 1-7 A cationic or neutral phase transition catalyst is preferably used in 5 - 30% of the weight of an amount one to 50% of the weight to the capacity of a formalin solution. Or it is the approach characterized by being preferably used in 3 - 10% of the weight of an amount 1 to 20% of the weight to the capacity of the water solution of a solid-state paraformaldehyde.

9. It is the approach that the mole ratio of 2for ****-ethyl hexanal is characterized by 1:2-4, and being 1:2.5-3.5 preferably in the process of said claims 1 or 7 at the formaldehyde used as an initial raw material.

10. It is the approach that an alkali-metal hydroxide or an alkaline earth metal hydroxide is characterized by 1-2-mol 1.25 to 1.75-mol twice and a thing [twice] to 2-ethyl hexanal in the process of said claims 1 or 6, preferably.

[Translation done.]

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

The manufacture approach of 2-n-butyl-2-ethyl-1,3-propanediol The purpose of this invention is 2-n-butyl-2-ethyl from 2-ethyl hexanal and a PORUMU aldehyde. — Newly for manufacturing I and 3-propanediol, and technically, it is easy and, moreover, is in offering an efficient process. This invention sets a foundation to the source of an initial component chosen correctly, and use of the catalyst chosen as accuracy.

2-n-butyl-2-ethyl — I and 3-propanediol are the compounds originally known well, as it is used for manufacture of polyester or is used for coating industry as one component of powder coatings. The advantage of this compound is with the UV electric shielding nature which was very excellent, and adsorbent [over the water / very small].

Originally manufacturing 2-n-butyl-2-ethyl-1,3-propanediol by the so-called Cannizzaro's reaction which follows aldol addition and it immediately is known in organic chemistry from 2-ethyl hexanal and formaldehyde. This reaction is attained by using a basic alkali-metal hydroxide catalyst or an alkaline-earth-metal hydroxide catalyst during a reaction.

The Japanese patent public presentation JP 48-043085 indicates manufacturing 2-n-butyl-2-ethyl-1 and 3-pro van diol by two staircase reactions under existence of a basic alkali-metal hydroxide from 2-ethyl hexanal and formaldehyde, and alternative aldol condensation is 30-60 degrees C, and it is performed by pH 8-11 in the 1st step. In the 2nd step, the alternative crossover-Cannizzaro's reaction is performed by pH 8-11 at 65-90 degrees C. However, the yield of about [that isolation of the 2-n-butyl-2-ethyl-1,3-propanediol obtained as an end product and the approach of purification are not reported] and an end product is not reported, either.

The Japanese patent public presentation JP 62-129233 indicates the approach for composition of 2-n-butyl-2-ethyl-1,3-propanediol, and purification: In this approach, it is obtained as a result of the reaction which obtains 2-n-butyl-2-ethyl-1,3-propanediol from 2-ethyl hexanal and formaldehyde under existence of an alkali-metal hydroxide or an alkaline earth metal hydroxide, and water. and it is neutralized by the content **** reaction mixture, the organic acid, or the mineral acid, and 2-n-butyl-2-ethyl-1,3-propanediol is separated in an organic layer — having — the water of the per [0.01] an organic layer or its section — 2 section — 1 time — or it is washed several times and formaldehyde is removed. Vacuum distillation of the organic layer is carried out to the last. In this process, when the starting material used is a paraformaldehyde, it is reported that the yield of the last purification object is 91.9%.

In the Japanese patent public presentation JP 2-062836, when the yield and grade of 2-n-butyl-2-ethyl-1,3-propanediol add C14 alcohol beyond 2 % of the weight or it to 2-ethyl hexanal in a heterogeneous system under existence of an alkali-metal hydroxide into the mixture of 2-ethyl hexanal and formaldehyde, it has been improved. An organic layer is separated from the mixture neutralized by the organic acid or the mineral acid, and pH of the organic layer is adjusted to 4.5-5.5 by phosphoric acid before the vacuum distillation. In the example, the reaction mixture contained 7.9% of the weight of the methanol to 2-ethyl hexanal, and it was reported that the yield of 2-n-butyl-2-ethyl-1,3-propanediol was 94.4% in this case.

At U.S. Pat. No. 2,413,803, 2-n-butyl-2-ethyl-1,3-propanediol is compounded by 2-ethyl hexanal

and formaldehyde to hydroxylation potash at 73% of yield through the synthetic long process by the multistage time from the homogeneous mixture equalized by ethanol.

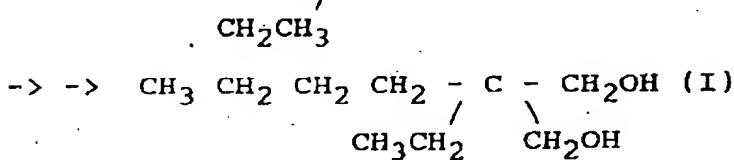
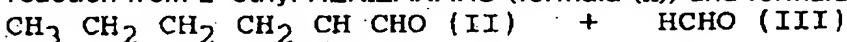
In U.S. Pat. No. 5,146,004, 2-ethyl-2-(hydroxymethyl) hexanal is manufactured from 2-ethyl hexanal, formaldehyde, and the 4th class amine. Any unreacted 2-ethyl hexanal is made [being isolated by carrying out azeotropic distillation of the organic layer in a rough product, or], and 2-butyl-2-ethyl-1,3-propanediol is obtained from the organic layer in which the rough product obtained by catalytic hydrogenation was refined at 85 – 88% of yield. Hydrogenation is performed by using a nickel catalyst.

In the process which is a general Prior art for all the processes by the official report by which reference was carried out [above-mentioned], and 2-n-butyl-2-ethyl-1,3-propanediol, continuation of a troublesome reaction is adopted in the multistage time. Entire high yield which was reported for the reported yield to be low or to base it on the experiment which is not reported at all or was carried out by relation with this invention was never attained except for the experimental result adjusted according to the Japanese patent public presentation JP 2-062836.

The purpose of the approach of this invention is in a thing [a thing] an end product can be obtained with yield very high as a result and which manufacture 2-n-butyl-2-ethyl-1,3-propanediol according to a simple and efficient process technically. That this purpose chooses correctly the source of a component kept for a reaction and by using the combination of a catalyst chosen correctly showed being attained at the surprising thing.

From 2-ethyl hexanal and formaldehyde, the approach of this invention of manufacturing 2-n-butyl-2-ethyl-1,3-propanediol is characterized according to the matter stated in the description partial knot in claim 1 by using an alkali-metal hydroxide catalyst or an alkali top metal hydroxide.

According to this invention, the 2-n-butyl-2-ethyl-1,3-propanediol shown by the formula (I) is compounded by using the so-called cationic or neutral phase transition catalyst for increasing efficiently an alkali-metal hydroxide catalyst or an alkaline earth metal hydroxide catalyst, and a reaction from 2-ethyl HEKIZANARU (formula (II)) and formaldehyde (formula (III)).



After a reaction process, phase separation of the mixture is neutralized and carried out by the organic acid or the mineral acid, an organic phase is washed with water and vacuum distillation is carried out.

It is new about a catalyst, and moreover, in order to advance a desired reaction combining surprising language especially said catalyst, i.e., alkali-metal hydroxide catalyst, or alkaline earth metal hydroxide catalyst + cationicity or a neutral phase transition catalyst in this invention and to make it increased, it is using it. Furthermore, as for the source of formaldehyde used, it is essential in less than 2% of methanol a content **** methanol free-lancer's formalin solution or that it is a solid paraformaldehyde again in this invention.

In a polyphase system, although the manufacture process indicated by this invention is a polyphase system, the internal phase transition of the matter is a factor which often controls a reaction rate. A reaction rate and reaction effectiveness can be increased thru/or raised by using the so-called chemistry phase transition catalyst by [which increase the internal phase transition of the matter] increasing mechanical agitation effectiveness so that it may depend especially and/or a remarkable internal phase contact front face may be generated. Chemical phase ***** is a chemical and the property is carrying out the catalyst of the internal phase transition of the matter. Phase transition catalysts may be cationicity and anionic and neutral any.

In the process of this invention, phases at present are an organic phase and aqueous phase. In this case, now, it was admitted that the advantageous effectiveness and the advantageous rate of the final result, i.e., a reaction, are attained by using the phase transition catalyst of neutrality or KARAON nature with an alkali-metal hydroxide or an alkaline-earth-metal hydroxide. It can do [that reference is made as a polyethylene glycol and a neutral phase transition catalyst, or].

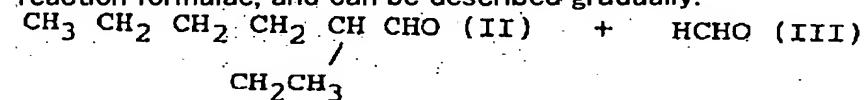
The example of a type of a cationic phase transition catalyst contains sulfuric-acid acidity tetrabutylammonium, TBAHS04, and a TORIKA prill methylammonium hydrochloride.

The functional principle of most cationic phase transition catalysts between an organic phase and a tank can be described as follows, using TBAHS04 as an example. underwater big cation TBA⁺ and underwater big anion HSO₄⁻ (or H⁺ and SO₄²⁻) — ion — pair twinning — it is emitted from a sulfuric-acid acidity tetrabutylammonium molecule. if this cation contacts the anion (namely, OH⁻) of monovalence, a new ion pair will form — having — that structure — ** — high (fat). formed ** — for high structure (fat structure), this new ion pair shifts to an organic phase simply and quickly. If an ion pair exists in an organic phase, the anion will react with a desired reactant. In this case, NaOH reacts with 2-ethylhexanol. Since OH⁻ carried by the ion pair reacts with an organic compound, TBA⁺ remains to an organic phase in the form of ion, and is disadvantageous. So, gammaBA⁺ returns to the aqueous phase. In the aqueous phase, the above-mentioned process in which a hydroxyl group is incorporated to an organic phase happens again.

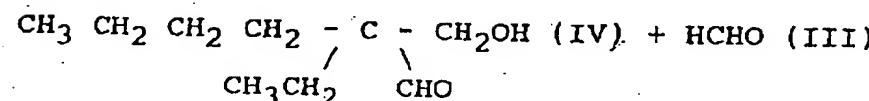
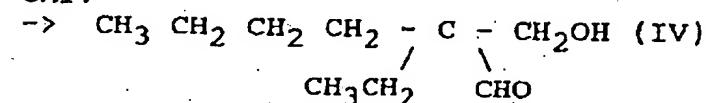
The operation principle of a neutral phase transition catalyst can be described as follows, using a polyethylene glycol as an example. The polyethylene glycol (PEG) does not have the charge. So, the operation is different from it of the cationic phase transition catalyst mentioned above. PEG acts as a phase transition catalyst combining with a cation, and by forming the so-called crown ether ****. The isolation electron of the oxygen atom in PEG draws a cation. The crown ether formed on account of the structure of PEG shifts into an organic phase. In order to attain an electronic equilibrium in a phase, an anion also shifts into an organic phase. For example, when a sodium hydroxide is used, crown ether is formed by Na⁺, it shifts to an organic phase, and PEG follows in footsteps, in order that OH⁻ may also form an electronic equilibrium. Since a hydroxide ion is consumed by the reaction produced in an organic phase, it is repeated in order that the above-mentioned cycle may attain an electronic equilibrium.

According to statement of the conventional technique, the chemistry phase transition catalyst was not used for manufacture of 2-n-butyl-2-ECHIRU 1,3-propanediol. Using using them with a basic catalyst and the reaction component chosen still more correctly constitutes the new approach for manufacturing said compound by advantageous and efficient technique about a manufacturing technology in the manufacture process of this invention.

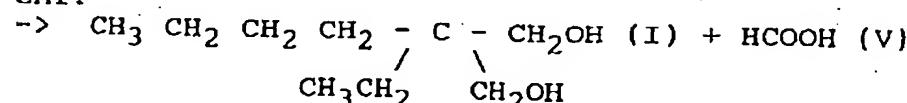
The chemical reaction produced in the manufacture process of this invention has the following reaction formulae, and can be described gradually.



CAT.



CAT.



Thus, it is the first reaction step and is 2-ethyl hexanal (formula (II)). And formaldehyde (formula (III)) reacts, it is an intermediate product and beta-hydroxyaldehyde, i.e., 2-n-butyl-2-ethyl-3-hydroxy propanal, shown by Formula IV is formed. In the viewpoint of the economical efficiency of a process, the Cannizzaro's reaction starts quickly after this aldol reaction between the first raw materials, and it sets to that Cannizzaro's reaction. Oxidize in a part of formaldehyde of a formula (III), and it becomes considerable ***** (formula V), i.e., formic acid. Becoming alcoholic ***** 2-n-butyl-2-ethyl-1,3-propanediol (formula I) which is returned and corresponds beta-hydroxyaldehyde (formula IV) of an intermediate product. But since it is important, the catalysts used for a reaction must be either an alkaline-earth-metal hydroxide and an alkali-metal hydroxide.

beta-hydroxyaldehyde, i.e., 2-n-butyl-2-ethyl-3-hydroxy propanal, whose object of this invention is an intermediate product is formed as quickly as possible and completely. It is returned as completely as quick moreover possible, and it becomes 2-n-butyl-2-ethyl-1,3-propanediol. And it is that an alkali-metal salt or an alkaline-earth-metal salt is further formed according to the formaldehyde superfluously used during the reaction, or the catalyst which oxidizes, becomes formic acid and is used. It can be attained that returning this object, i.e., beta-hydroxyaldehyde, to forming completely to the maximum extent and alcohol chooses a proper reaction condition and the refined initial raw material component (especially it is applied about formaldehyde), and by using a neutral or cationic phase transition catalyst additionally in this invention. By these, the uneven system of reaction in the first stage becomes homogeneity further, a reaction advances to still quicker and much more advantageous extent, and it becomes so high that the yield of desired 2-n-butyl-2-ethyl-1,3-propanediol is surprised as the final result moreover obtained.

It is essential again that the source of the formaldehyde used as an initial source material is correctly chosen for an efficient and economical reaction. The source of formaldehyde where this demand is used is filled, a paraformaldehyde (more efficient use of a capacity), or when it is almost 30 - 53% of the weight of a methanol free-lancer's (MeOH<2 % of the weight) formalin solution. (The most economical thing is using the formalin solution directly obtained, for example in this process.)

The mole ratio to the formaldehyde of 2-ethyl hexanal used as an initial raw material of a reaction is within the limits of 1:2-4, and is 1:2.5-3 preferably.

It should be within the limits of 5. A neutral or cationic phase transition catalyst (for example, PEG400), for example, a neutral polyethylene glycol, the cationic 4th class ammonium (TBAHSO₄) of sulfuric-acid acidity, or a TORIKA prill methylammonium hydrochloride (Aliquat 336) is added by the mixture. It is preferably added in 3 - 10% of amount one to 20% of the weight, or these are preferably added [as opposed to / when used in a solid paraformaldehyde by being 1 - 50% of the weight of an amount, and being preferably added in 5 - 30% of the weight of an amount to the capacity of a formalin solution / the capacity of the water solution of a solid paraformaldehyde] at 5 - 15 % of the weight one to 50% of the weight per amount of 2-ethyl hexanal. The mixture of an initial raw material is heated most preferably [it is desirable and] at 50-80 degrees C by 30-100 degrees C at 60-80 degrees C. And dropping addition of the water solution of an alkali-metal hydroxide or an alkaline-earth-metal hydroxide is preferably carried out over 1 - 3 hours for 0 to 5 hours at this mixture. the reaction — said temperature — 0 - 3 hours — it is continued still more preferably for 15 minutes to 15 minutes per hour for 15 minutes to 2 hours. an alkali-metal hydroxide or an alkaline earth metal hydroxide — 1 - 2 double molar quantity of 2-ethyl hexanal — it is preferably used by molar quantity 1.25 to 1.75 times. Behind this reaction phase, said mixture is an organic acid or a mineral acid, and is preferably neutralized by the sulfuric acid. And it is classified mutually, and a phase is mutually classified, while [desirable] it is hot. an organic phase — 1 time — or several times, it is water of at least 50% of capacity of the capacity of an organic phase, and is washed one to twice preferably. A phase is separated and vacuum distillation is carried out together with an organic phase. The boiling points of an end product are 130-132 degrees C / 9mmHg. Although the yield of the 2-n-butyl-2-ethyl-1,3-propanediol obtained is based also on a reaction condition and a

mole ratio, it is 92% or more.

The suitable mode attained using the cationic or neutral phase transition catalyst used in the process of this invention is determined according to the used source of formaldehyde. If the source of formaldehyde used is a content **** formalin solution about a methanol at less than 1% preferably less than 2%, a cationic and neutral phase transition catalyst is a suitable phase transition catalyst for bringing about the advantageous final result. Supposing a solid paraformaldehyde is used as a source of formaldehyde, the above-mentioned 4th class ammonium of sulfuric-acid acidity or a TORIKA prill methyl ANMMONIUMU hydrochloride is suitable to attain the highest possible yield.

It is simple, and the approach of this invention of manufacturing the 2-n-butyl-2-ethyl-1,3-propanediol described above is reliable, is efficient, and is an approach of manufacturing this compound by high yield.

The manufacture process by this invention is illustrated very much to a detail in the following examples.

However, they are only for illustrating this invention and must not understand this invention restrictively.

<The example for which the formalin solution was used as a source of formaldehyde> (Example 1)

It was inserted in in the reactor, 136.3g 45% formalin solution containing less than 1% of the weight of a methanol, 100.5g 2-ethyl hexanal, and 13.8g neutral (it is 10 % of the weight to the capacity of a formalin solution) a phase transition catalyst (PEG400), i.e., BORIECHIREN glucohol. And mixture was heated by 70 degrees C. The 142.0g NaOH water solution was added in this mixture over 2 hours. The reaction was continued for 45 minutes. This mixture was neutralized by the sulfuric acid. The phase was separated. It was washed twice by an organic phase or water. Vacuum distillation of the end product was carried out by 130-132 degrees C / 9mmHg. The 116.5g 2-n-butyl-2-ethyl-1,3-propanediol which is equivalent to 92.7% of yield calculated from 2-ethyl hexanal with this distilled off.

(Example 2)

The reaction was carried out like said example 1 by the outside where the polyethylene glycol was used at 30% of the weight of a rate to the capacity of a formalin solution. 2-n-butyl-2-ethyl-1,3-propanediol had and distilled off 94.2% of yield.

(Example 3)

The reaction was carried out like said example 1 by the outside where the polyethylene glycol was used at 3% of the weight of a rate to the capacity of a formalin solution. 2-n-butyl-2-ethyl-1,3-propanediol had and distilled off 84.1% of yield.

(Example 4)

Outside where the polyethylene glycol was used at 1% of the weight of a mere rate to the capacity of a formalin solution was carried out in the reaction like said example 1. 2-n-butyl-2-ethyl-1,3-propanediol had and distilled off 76.7% of yield.

(Example 5)

It was heated by 60 degrees C, 69.8g 45% formalin solution containing less than 1% of the weight of a methanol, 50.2g 2-ethyl hexanal, and 4.1g cationic (it is 5.9 % of the weight to the capacity of a formalin solution) the phase transition catalyst 336, i.e., Aliquat. The 71.0g NaOH water solution was added in this solution over 2 hours. Mixture was heated for 45 minutes. After that, mixture was neutralized by the sulfuric acid. The phase was separated. The organic phase was washed once by water. Vacuum distillation of the 55.7g 2-n-butyl-2-ethyl-1,3-propanediol was carried out by 130-132 degrees C / 9mmHg. Under [this is equivalent to 88.8% of yield].

(Example 6)

The reaction was carried out like said example 5 by the outside where Aliquat 336 was transposed to the 4th class ammonium of sulfuric-acid acidity, and TBAHSO4, and was used at 6.0% of the weight per capacity of it or a formalin solution of a rate. 2-n-butyl-2-ethyl-1,3-propanediol and 84.6% of yield was had and distilled off.

(Example 7)

It was heated by 70 degrees C, 134.0g 45% formalin solution containing less than 1% of the weight

of a methanol, 100.6g 2-ethyl hexanal, and 13.3g cationic (it is 10 % of the weight to the capacity of a formalin solution) the phase transition catalyst 336; i.e., Aliquat. The 142g NaOH water solution was added in this solution over 2 hours and 20 minutes. It was agitated for 45 minutes in mixture. After that, mixture was neutralized by the sulfuric acid. The phase was separated. An organic phase is 1-time washing ***** with water. Vacuum distillation of the 117.0g 2-n-butyl-2-ethyl-1,3-propanediol was carried out by 130-132 degrees C / 9mmHg. Under [this is equivalent to 93.0% of yield].

(Example 8)

The reaction was carried out like said example 7 by the outside where Aliquat 336 was transposed to the 4th class ammonium of sulfuric-acid acidity, and TBAHSO4, and was used at 10% of the weight per capacity of it or a formalin solution of a rate. 2-n-butyl-2-ethyl-1,3-propanediol had and distilled off 87.8% of yield.

Example 9: (— the example of a comparison

Reaction execution of the outside where a phase transition catalyst was not used during the reaction was carried out like said example 1. 2-n-butyl-2-ethyl-1,3-propanediol had and distilled off 76.5% of yield.

<The example for which the solid-state paraformaldehyde was used as a source of formaldehyde> (Example 10)

A 29.9g solid-state paraformaldehyde, 50g water, 50.0g 2-ethyl hexanal, and 4.1g cationic (it is 5.1 % of the weight to the capacity of the water solution of a paraformaldehyde) phase transition catalyst ***** Aliquat 336 were heated by 60 degrees C. The 125.0g NaOH water solution was added in this solution over 1.5 hours. Mixture was agitated further for 1.5 hours. After that, mixture was neutralized by the sulfuric acid. The phase was separated. The organic phase was washed twice by water. In 130-132 degrees C / 9mmHg, vacuum distillation of the 56.9g 2-n-butyl-2-ethyl-1,3-propanediol was carried out, and it was 91.0% of yield.

(Example 11)

29.9g solid-state rose formaldehyde, 50g water, 50.2g 2-ethyl hexanal, and 4.1g TBAHSO (it is 5.2 % of the weight to the capacity of the water solution of a paraformaldehyde)4 were heated by 60 degrees C. The 125.0g NaOH water solution was added in this solution over 2 hours. Mixture was agitated further for 1 hour. After that, mixture was neutralized by the sulfuric acid. The phase was separated. The organic phase was washed twice with water. In 130-132 degrees C / 9mmHg, vacuum distillation of the 55.0g 2-n-butyl-2-ethyl-1,3-propanediol was carried out, and it was 87.7% of yield.

(Example 12)

The reaction was carried out like said example 11 by the outside where the polyethylene glycol which is a neutral phase transition catalyst was used instead of the cationic phase transition catalyst in the amount of 4.0g (it is 4.9 % of the weight to the capacity of the water solution of a paraformaldehyde). The yield of the obtained 2-n-butyl-2-ethyl-1,3-propanediol was 73.2%.

(Example 13; example of a comparison)

The reaction was carried out like said example 9 by the outside where a phase transition catalyst was not used.

The yield of the obtained 2-n-butyl-2-ethyl-1,3-propanediol was 73.2%.

According to the above-mentioned example, when a cationic or neutral phase transition catalyst is used, it is clear that yield quite better than the case where these catalysts are not used is obtained. Furthermore, in a suitable mode, if the source of formaldehyde is almost a methanol free-lancer's formalin solution, it is possible to use both cationic and neutral phase transition catalysts, but if a solid paraformaldehyde is used, it will be desirable to use a cationic phase transition catalyst.

[Translation done.]